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Frequency Response of Electrical Properties of some Granite Samples

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Abstract

The electrical properties of rock types or channel structures have been the object of intense studies for many years. The main aim of the present work is to show the frequency response of electrical properties of some Granite samples concerning chemical, minor and major composition. This may be used as a fingerprint for the characterization of some Granite rocks. Electrical measurements on Granite samples (surface, Aswan, Egypt) were measured (100 Hz up to 10^7 Hz). Differences in samples due to changes in texture lead to different changes in electrical properties. The changes at conductivity and dielectric constant are reflections from the texture within grains. Conductivity and dielectric constant values extend from $\sim10^{-8}$ to 10^{-4} (S/m) and 24 to 3.5, respectively. With frequency increase, conductivity increases, and dielectric constant decreases (10 kHz), and then it settles down. Differences in electrical properties between samples are small as a direct result of the homogeneity and texture of most samples. There is a high dispersion of dielectric constant for relatively low-frequency values (<10 kHz) and no dispersion at relatively high frequencies. In conclusion, the dielectric constant decreases due to the shrinkage of energy levels for electrons and the increase of hopping particles between different particles.

Keywords: Conductivity; Granite; Frequency-domain; Electric; Dielectric constant.

1. Introduction

Minor contributions to the electrical properties are provided by pressure, bound water, oxygen fugacity, and other parameters. Electrical properties of the studied samples and related rock types or channel structures have been the object of intense studies for many years (Gomaa, 2020a). Granite is supposed to be an igneous plutonic rock. It has roughly equal quantities of the sodic plagioclase, potassium feldspar, and quartz. Granites color changes according to their mineralogy and chemistry from black to dark gray or pink. Granite does not have internal structures, and it is hard and may be used for construction. Granite may be granitoid (coarse-grained plutonic rocks). The name of the rock comes from the alkali feldspar concentration (microcline, orthoclase, or sanidine), quartz percentage, and plagioclase feldspar. True Granite may contain both alkali feldspars and plagioclase. The primary permeability of Granite is very poor with very strong secondary (not examined in our case) permeability (Charlier et al., 2011, Namur et al., 2011, Kassab et al., 2017). Figure (1) shows (A) Fine-grained Granite; (B) Porphyritic Granite showing large alkali-feldspar grains in a fine-grained matrix.

a. Chemical and mineralogical composition of Granite rock: The morphology of the grains is randomly oriented with a spheroidal shape. Some samples (surface, Aswan, Egypt) may have small subhedral grains alkali-feldspar (Figure 1). Quartz percentage is in the order of 30% (Table 1, Table 2). Antiperthitic zoned plagioclase is found with a concentration of less than 50%. Quartz is found with a concentration of ~10% (Leake et al., 1997; Brent et al., 2010; Wang et al., 2006; Yoshino et al., 2006). The average chemical composition of Granite is SiO_2 (72.04%), Al_2O_3 (14.42%), K_2O (4.12%), Na_2O (3.69%), CaO (1.82%), FeO (1.68%), Fe_2O_3 (1.22%), MgO (0.71%), TiO_2 (0.30%), P_2O_5 (0.12%), and MnO (0.05%) (Geology Science, 2020).

b. Previous work on Granite rocks: Manghnani and Rai (1978) search for the effects of H_2O on the electrical conductivity and its anisotropy in olivine (Fo_{90}) at 8 GPa using complex impedance spectroscopy. They found that the increasing of H_2O content increases the conductivity, but activation energies are lower and H_2O concentration-dependent. Wang et al. (2006) measured hydrous polycrystalline olivine conductivity at 4 GPa and concluded that 80

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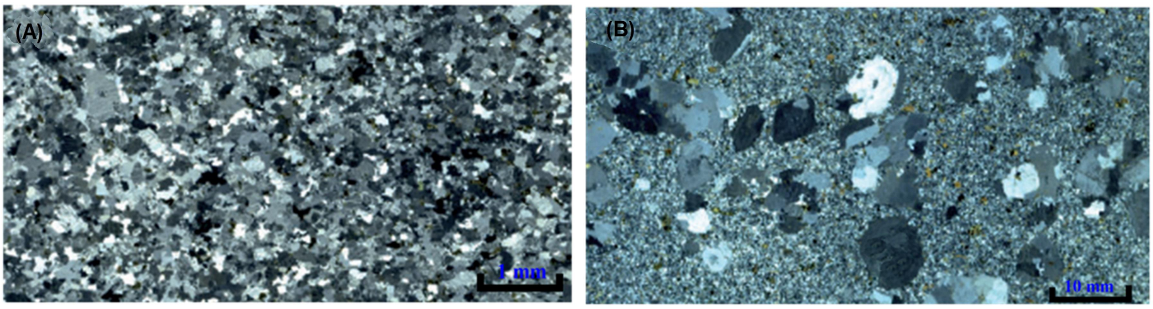
wt ppm H_2O in olivine would increase its conductivity by more than a factor of 10 at 1400 ◦C and account for the generally high conductivity of the oceanic asthenosphere. Yoshino et al. (2006), could not define how the conductivity changes with changing water (concentration) content along with all crystallographic directions, but rather assumed that conductivity increased linearly with H_2O concentration without change in activation energy (Watanabe, 1970; Presnall et al., 1972; Murase and McBirney, 1973; Rai and Manghnani, 1976; Shankland and Waff, 1977).



(a) (b)

Figure 1. Shows (a) Fine-grained Granite; (b) Porphyritic Granite showing large alkali-feldspar grains in a fine-grained matrix.

Table 1. Shows minerals in some silicic samples of Granite.

Rock-type	Feldspars	Quartz	FeTiOx	Cpx	Opx	Ol	Amph	Ap
Granite	64·6	32·8	0·3	–	–	–	3·0	0·3
	63·8	27·6	0·8	–	–	–	7·6	0·2
	61·2	33·6	0·6	–	–	–	14·2	0·3
	65·5	19·4	1·2	0·3	–	–	12·7	0·4
	64·5	21·4	0·9	–	–	–		0 5

Table 2. Shows major element compositions (wt%) of Granite.

SiO_2	Al_2O_3	FeO_t	CaO	Na_2O	K_2O	Total	An*	Any+	Abz++	Or§
68	19	0	0	9	1	100	0	0	91	8
67	20	0	1	10	0	100	4	4	92	3
67	20	0	0	10	1	99	1	1	90	8
68	20	0	0	11	0	100	2	2	96	0
68	19	0	0	11	0	100	0	0	97	2
67	19	0	0	10	0	99	0	0	96	3
68	19	0	0	10	1	100	0	0	93	6
65	18	0	0	1	14	100	0	0	10	89
64	18	0	0	1	14	99	1	0	15	84
64	18	0	0	1	14	99	5	0	12	87
65	18	0	0	1	14	99	0	0	10	89
64	18	0	0	0	15	99	0	0	8	92
65	18	0	0	1	13	99	0	0	15	84
65	18	0	0	0	15	100	0	0	7	92

$FeO_t \rightarrow FeO_{total}$, *An¼100[Ca/(CaNa)],+An¼100[Ca/(CaNaK)], ++Ab¼100[Na/(CaNaK)], §Or¼100[K/(CaNaK)].

Manghnani (1978) investigated the electrical conductivity of two ultramafic rocks (lherzolite and peridotite) to melting temperature at 1 bar under a certain oxygen fugacity environment. The electrical conductivity was found to increase with the degree of partial melting. Also, they found that there is a certain necessary melt fraction (~ 15 %) for the electrical conductivity to increase by nearly one order of magnitude. The electrical conductivity of the two rocks increases slowly until the formation of ~ 15 % melt fraction. Above that concentration (this is a critical concentration), the conductivity increases rapidly and monotonically. For a certain melt fraction, the electrical conductivity of lherzolite is lower than that of peridotite. This was attributed to the differences in the formed composition of the two melts. The conductivity of partially molten rocks depends on the concentration of melt and the degree of wetting of the grain boundaries by the melt (Waff, 1974).

Basalts laboratory measurements under temperatures (at 1 atm) of electrical conductivity show that through the melting interval, the conductivity of basalts increases from 1 to 2 orders of magnitude (Watnabe, 1970; Presnall et al., 1972; Murase and McBirney, 1973; Rai and Manghnani, 1976). The conductivity of basaltic melts is about two to three orders of magnitude greater than the electrical conductivity of olivine in the (1200 ^{0}- 1500 ^{0}C). The electrical conductivity increases, systematically, with increasing clinopyroxene, garnet, and alkalis (Na_2O + K_2O). These minerals are the major contributors of the melt formation. The changes of the ultramafic rocks of electrical conductivity values were attributed to the variations in the degree of partial melting.

Olhoeft (1981) shows that the electrical properties of Granite are controlled by the amount of free water and temperature. The effect of sulfur fugacity may be important but is experimentally unconfirmed. Also, changing the chemistry of water in Granite changes the temperature dependence of the electrical properties. With the increase of temperature, changes in water content are large and higher than the temperature (i.e. the water effect is higher than the temperature effect). At room temperature, the first water monolayer will increase the electrical conductivity. With more water monolayers, the electrical conductivity increase by ~ 9 orders of the magnitude and decrease the thermal activation energy (~ 5). Below the melting point of Granite (650^0 to 1100^0 C, according to the water pressure) some concentrations of water increase the conductivity with ~ 3 orders of magnitude and the activation energy by a factor of 2. Above the melting point of Granite (650^0 to 1100^0 C, according to water pressure) there are small concentrations of water that will increase the conductivity by <1 of magnitude and will barely change the activation energy. Measured samples of hydrated hornblende schist (with structural water) show an electrical conductivity comparable to the electrical measurements for dry Granite. The combinations of all these results with laboratory electrical conductivity suggest that the presence of water is important for the behavior of electricity (Gomaa, 2009; Gomaa and Gobara, 2020).

The existence of water vapor (adsorbed at the surface area of the insulator, e.g. basalt) has a very minute effect on electrical properties (this may be called the humidity, Gomaa and Eldiwany, 2020). Effect of water will be still very minute until a sufficient amount of water (~20% of a monolayer or ~ 0.002 %) is present between the pores to produce a connected path through the pore system (Olhoeft, 1981; Gomaa, 2008). One monolayer is required for the dielectric constant to begin to change; at the same time, the DC conductivity will be increased by nearly one order of magnitude (Olhoeft, 1981; Gomaa, 2008). The increase of water weight percent increases the conductivity by nearly 9 orders of magnitude. The effect of oxygen fugacity on electrical conductivity is so small and can be neglected compared to the temperature and other parameters.

The amplitude of the electrical conductivity changes by many orders of magnitude with the additions of water to Granite, and with the temperature change. Without temperature, the activation energy of the electrical conductivity of dry Granite is in the order of ~0.5 eV and increases to ~1.5 eV with the increase of the temperature. The activation energies in wet Granite are near 0.1 eV and very constant with increasing temperature.

The conductivity of the rocks depends on the capacity of the rock to absorb water and the kind and texture (structure of the pore spaces), the amount and composition of the retained water between pores (Gomaa and Kenawy, 2020). Hence, the conductivity may depend, to an important extent, upon the formation mechanism of the geological and climatic environment in which the material happens to find itself (Louis et al., 1942). In general, the conductivity is strongly dependent upon the direction, for anisotropic rocks (e.g. for schistose the conductivity in the direction of bedding is nearly 100 times greater than the conductivity in the perpendicular direction to bedding) (Gomaa, 2020a-e). Recently, the effect of variations in composition upon the electrical conductivity of artificially prepared samples of sulfides and oxides was measured. It was found that a slight excess of the free metal may increase the conductivity in some instances by a thousand fold. Wide differences in conductivity commonly experienced in measurements upon natural massive specimens of the metallic sulfides are usually caused by relatively minor differences in composition. Also, the concentration needed to establish an effective continuous electrical path is dependent upon the type of the mineral concentration and the nature of the conducting minerals present.

Under a certain condition, minerals and rocks, the electrical conductivity of the laboratory measurement (surface samples) may help us to make a reasonable explanation for available results of underground resistivity methods to provide much information about the chemical composition, mineral composition, and phase state of materials in the deep interior of the earth, their texture and structure, and the thermal structure of the interior of the Earth (Dai et al., 2008). Furthermore, it is an important approach to understand the phase transition, partial melting, dehydration, diffusion, rheology, and energy transmission of rocks and soils, and how materials interact at the earth's deep interior.

The main reason responsible for the great dependence of the electrical conductivity on the frequency is that the different frequency ranges of the conduction behaviors are different for mineral and rock of grain interior, grain boundary, and sample-electrodes.

The main aim of the present work is to show the frequency response of electrical properties of some Granite samples concerning the chemical, minor and major composition.

2. Sample Preparation and Measurements

The general chemical composition (on average) of Granite may be found as SiO_2 (~70 %), Al_2O_3 (~15 %), K_2O (~4 %), Na_2O (~4 %), CaO (~2 %), FeO (~2%), Fe_2O_3 (~1 %) with other minor elements like MgO, TiO_2, P_2O_5, and MnO. Granite has roughly got quantities of sodic plagioclase, potassium feldspar, and quartz. They are hard and do not have internal structures. Primary permeability at Granite is nearly zero.

Electrical measurements were made on samples, of thin shape disks, with a diameter to thickness ratio of 5 to 1. A technique of two electrodes (Agilent dielectric test fixture 16451B) was used. The current density was ~1.1 to 1.2X 10^{-3} (μA/cm^2). Data were measured in the frequency range from 100 Hz up to 10^7 Hz at room temperature (~25^0 C) and a voltage of 1 V using a Hioki 3522-50 LCR Hitester Impedance Analyzer. Samples dimensions were chosen to avoid the fringing effect and other related (stray, capacitive …) effects. The samples used are dry and measured in an evacuated desiccator (Gomaa and Alikaj, 2009).

The impedance (Z) or the admittance (Y) can be used to characterize the electrical properties of the samples. The admittance $Y = G + jwC$, where G is the parallel conductance of the sample (Ohm^{-1}), $j = \sqrt{-1}$, $w = 2\pi f$ (f is the frequency), and C is the parallel capacitance of the sample (in Farad) (Olhoeft, 1981). The impedance Z is given by: $Z = R_s + \frac{1}{jwC_S}$, where R_S and C_s are the series resistance (in Ohm) and capacitance (in Farad), respectively. The complex conductivity $\sigma^* = \sigma' + j\sigma''$, where $\sigma' = w\varepsilon''$ and $\sigma'' = w\varepsilon'$, ε'' and ε' are the dielectric loss (energy that goes into heating a dielectric material in a varying electric field) and dielectric constant, respectively (Gomaa and Alikaj, 2009). Electrical properties are suggested to interpret the mechanisms of semiconductors and other rock processes (Gomaa et al., 2009).

The series and parallel capacitance and resistance were measured at different frequencies. The complex relative dielectric constant could be written as $\varepsilon^* = \varepsilon' - i\varepsilon''$, where the real part of the complex dielectric constant $\varepsilon' = C_p d / \varepsilon_0 A$ and the imaginary part $\varepsilon'' = G_p d / w\varepsilon_0 A$ is related to the measured parameters, A is the cross-sectional area of the sample, d is its thickness, ε_0 is the permittivity of free space (8.85×10^{-12} F/m), w is the angular frequency, G_p is the parallel conductance, and C_p is the parallel capacitance (Olhoeft, 1981).

3. Results and Discussion

Electrical measurements on the studied samples were carried out in the range of 100 Hz up to 10^7 Hz at room temperature (~25^0 C) and a voltage of 1 V. All the samples are electrically homogeneous (samples show the same behavior). The samples have a poor primary permeability. Samples grain size in the order of ~0·1 to 0·5 mm and the grains are randomly oriented (Charlier et al., 2011; Namur et al., 2011). The electrical conductivity changes from one rock type to the other, which is attributed to the differences in formation composition. The conductivity of partially molten rocks is much higher than that of solid rocks (Gomaa and Abou El-Anwar, 2015). The electrical properties of Granite change according to the amount of free water and temperature (Olhoeft, 1981; Gomaa, 2009). Water addition to samples makes the first water monolayer that increases the electrical conductivity. With more water monolayers, the electrical conductivity increases. The combinations of all these laboratory electrical conductivity measurements suggest that the humidity or the saturation of water is important for the behavior of electrical and dielectric measurements (Gomaa and ElSayed, 2006, 2009). Adsorbed water vapor at the surface area of the insulator, like Granite, has a minute effect on electrical properties. Effect of water will be still very minute until a connected path is present between the pores to obtain a connected pore system. Dielectric constant will begin to change from the moment of the presence of the first monolayer and the DC conductivity will be increased at the same time (Olhoeft, 1981; Abou El-Anwar and Gomaa, 2013; Gomaa, 2013). The increase of monolayers increases the conductivity monotonically while the dielectric constant will be increased up to the percolation threshold, then it will be decreased dramatically. The electrical conductivity changes by strikingly large amounts with the addition of water. The conductivity of the rocks depends on the amount and composition of the retained water between pores. Also, the conductivity may depend on the geological and climatic environment of the material origin and upon the direction of the bedding (Louis et al., 1942; Khater et al., 2019b; Khater et al., 2020; Gomaa, 2012; Shaltout et al., 2012). Minor differences in the composition may lead to a great change in the electrical conductivity.

Figure 2 shows the conductivity of eight Granite samples. From the behavior of the samples, there is one slope (0.75) for all the samples. There is no DC conductivity from the samples. The conductivity values at 100 Hz range from nearly $4\text{x}10^{-8}$ to $7\text{x}10^{-8}$ (S/m). The conductivity values at 5 MHz range from nearly $0.5\text{x}10^{-4}$ to $1.5\text{x}10^{-4}$ (S/m). Then the conductivity values extend from 10^{-8} to 10^{-4} (S/m). With the increase of the frequency, the conductivity increases. The conductivity values increase monotonically (Gomaa, 2006). The differences in the conductivity between the samples are small and they can be neglected. This is a clear indication that the samples are homogeneous and may have nearly the same composition (Table 1 and 2). The samples behavior obeys Jonscher's law ($\sigma \propto \mathrm{w}^{\mathrm{n}}, n \leq 1, w = 2\pi f$). There is no relation between the linearity and n. Table 1 shows concentrations of minerals in some silicic samples of Granite. Feldspars concentration is nearly 61 to 65%, and Quartz concentration is nearly 19 to 34%, FeTiOx concentration is nearly 0.3 to 1.2%, Cpx concentration is nearly 0 to 0.3 %, Amph concentration is nearly 3 to 14.2%, Ap concentration is nearly 0.2 to 0.5%. From the table of major element compositions (wt %) of Granite (Table 2), SiO_2 ranges from 64 up to 68%, Al_2O_3 ranges from 18 up to 20%, Na_2O ranges from 0 up to 11%, and K_2O ranges from 0 up to 14%.

Figure 3 shows the dielectric constant of Granite samples. From the behavior of the samples, there are two slopes for all the samples. The first slope (~0.25) is for the decrease of the dielectric constant with frequency for relatively low-frequency values. There is a high dispersion for these relatively low-frequency values. The other relatively high-frequency slope has nearly no dispersion (~0) of dielectric constant with the frequency. At relatively low-frequency values with the increase of frequency, the dielectric constant decreases due to the shrinkage of energy levels for electrons and the increase of the possibility of hopping between different particles. No flat area of dielectric constant at relatively low-frequency values, and accordingly, there are no DC links between particles at all the samples. The dielectric constant values range from nearly 24 at 100 Hz to 3.5 at 5 MHz. The dielectric constant values decrease up to nearly 10 kHz and then it settles down after that frequency. The differences in the dielectric constant values between samples are small that may be negligible. This indicates the homogeneity of the samples that nearly have the same composition with different ineffective texture variations (Abou El-Anwar and Gomaa, 2016; Gomaa and Kassab, 2016; Gomaa and Kassab, 2017). The samples behavior obeys Jonscher's law $(\epsilon \propto w^{1-n}, n \leq 1)$. This phenomenon can be interpreted by the transport of charge carriers. The increase of the frequency, which results in a smaller period of the voltage cycle, changes the transport and accumulation of charge carriers. The transport of charge carriers becomes difficult (at disconnected links) when the frequency is increased.

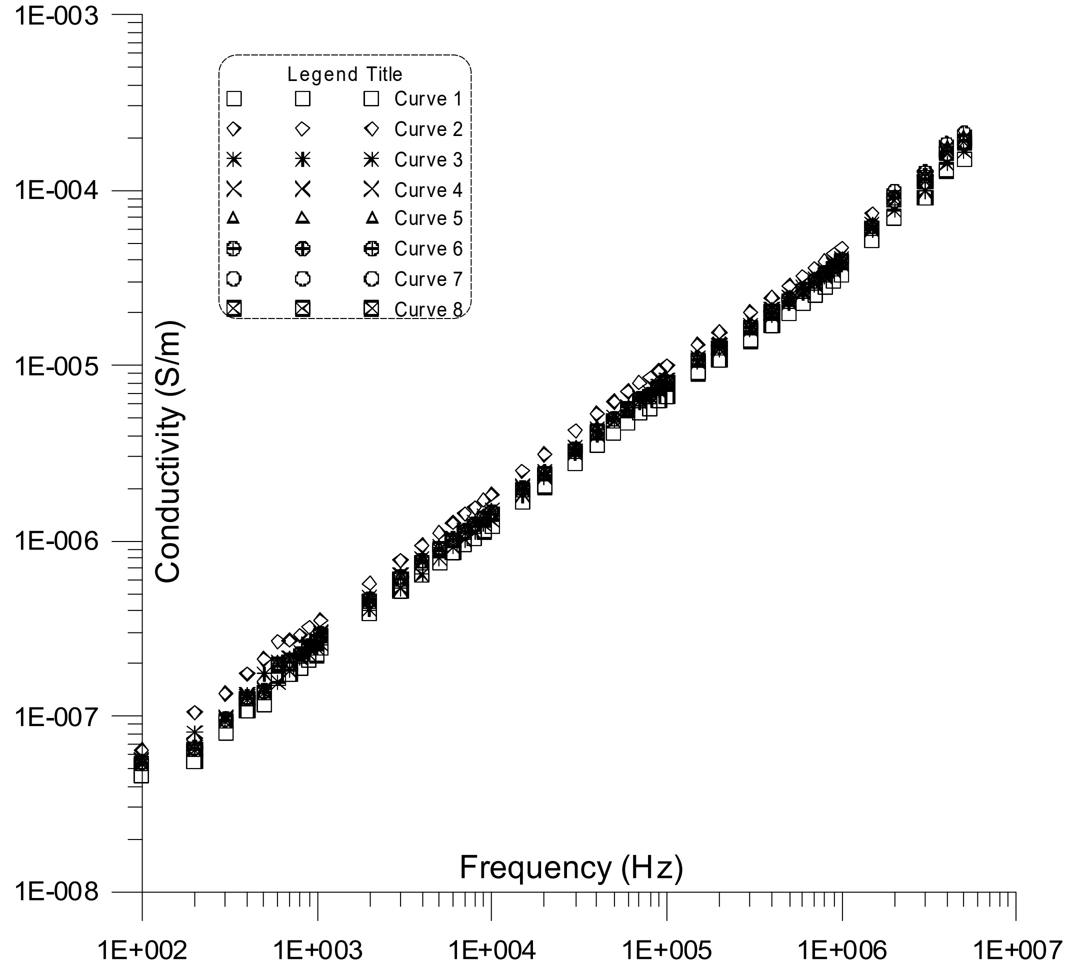


Figure 2. shows the conductivity of the eight Granite samples.

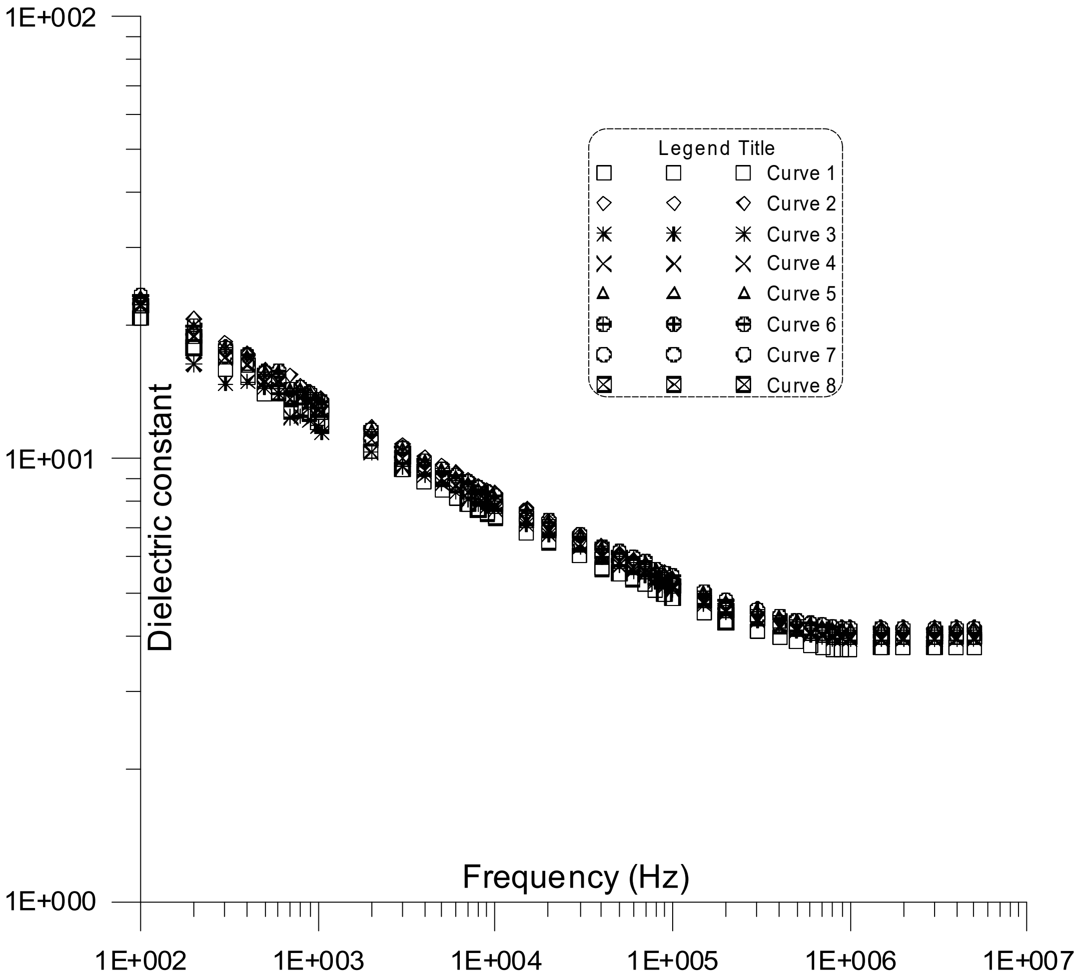


Figure 3. shows the dielectric constant of the eight Granite samples.

Figure 4 shows the dielectric loss (Tan *δ*) of Granite samples with the frequency. The dielectric losses in a solid dielectric material, under the influence of an alternating electric field, resulting from the motion of either permanent or induced dipoles in their effort to be aligned with the field or the motion of free charges between two or more possible equilibrium sites, separated by a potential barrier. From the behavior of the samples, there are two slopes for all the samples. The first slope is for relatively low-frequency values. There is a relatively low dispersion for these relatively low-frequency values (Gomaa et al., 2019a,b; Gomaa and Abou El-Anwar, 2019). The other relatively high-frequency slope has steep dispersion with the frequency.

At relatively low-frequency values with the increase of the frequency, the dielectric loss decreases due to the loss of energy as a result of the friction of free charges between equilibrium sites levels for electrons and the increase of hopping between different particles. The dielectric loss values range from nearly 0.5 at 100 Hz to 0.14 at 5 MHz. Again, the differences in the dielectric loss values between samples may be negligible, which indicates the homogeneity of samples (Gomaa et al., 2015a, b).

Figure (5) shows the complex impedance of Granite samples. From Figure 5, we can see the relation between the imaginary impedance at the Y-axis and the real impedance at the X-axis. From the behavior of the samples, there is a skewed arc (or semicircular arc) of the complex impedance. The representation of the complex impedance shows the variation of the reactance (capacitors and inductors) at the Y-axis with the resistance at the X-axis. The effect of reactance is similar to resistance. Reactance and resistance lead to smaller currents for the same applied voltage. Reactance is different from resistance, as reactance changes the phase (current is shifted by a quarter of a cycle relative to the voltage), and power is not dissipated in a purely reactive element but is stored instead. Also, reactance may cancel each other and have frequency dependence. Resistors have typically the same resistance for all frequencies. Mathematically, frequency (f) is taken as the parameter of the circumferential curve. The two endpoints, at $f = w/2\pi = 0$ and ∞, represent the intersection point of the impedance skewed arc and the X-axis, respectively. The point $f = 1/2\pi RC$ is just the highest point of the semicircle, where C is the capacitance and R is the resistance. The frequency gradually decreases from very high values to zero (Gomaa et al., 2018). From these curves, it is clear that the samples are insulators because the skewed arcs are near the Y-axis (resistance is very high). The present is a skewed arc (part of the semicircle) and was not a half-circle because the samples are resistive. The semicircle may be completed with the increase of frequency range, and then the arc will begin to be a semicircle. The differences of the complex impedance values are small between samples that may be negligible. This indicates the homogeneity of the samples (similar composition and small ineffective texture variations).

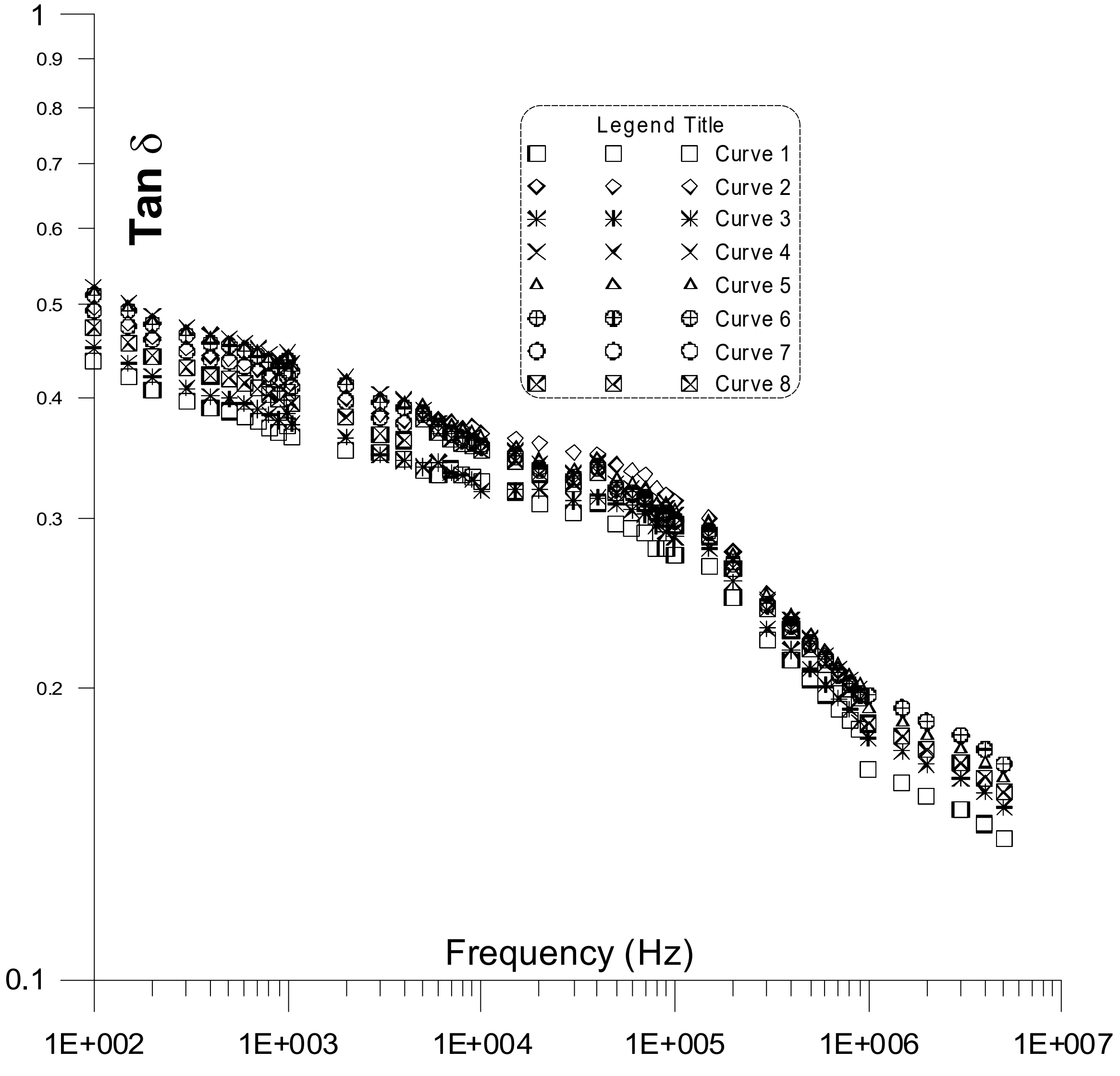


Figure 4. shows the dielectric loss of the eight Granite samples.

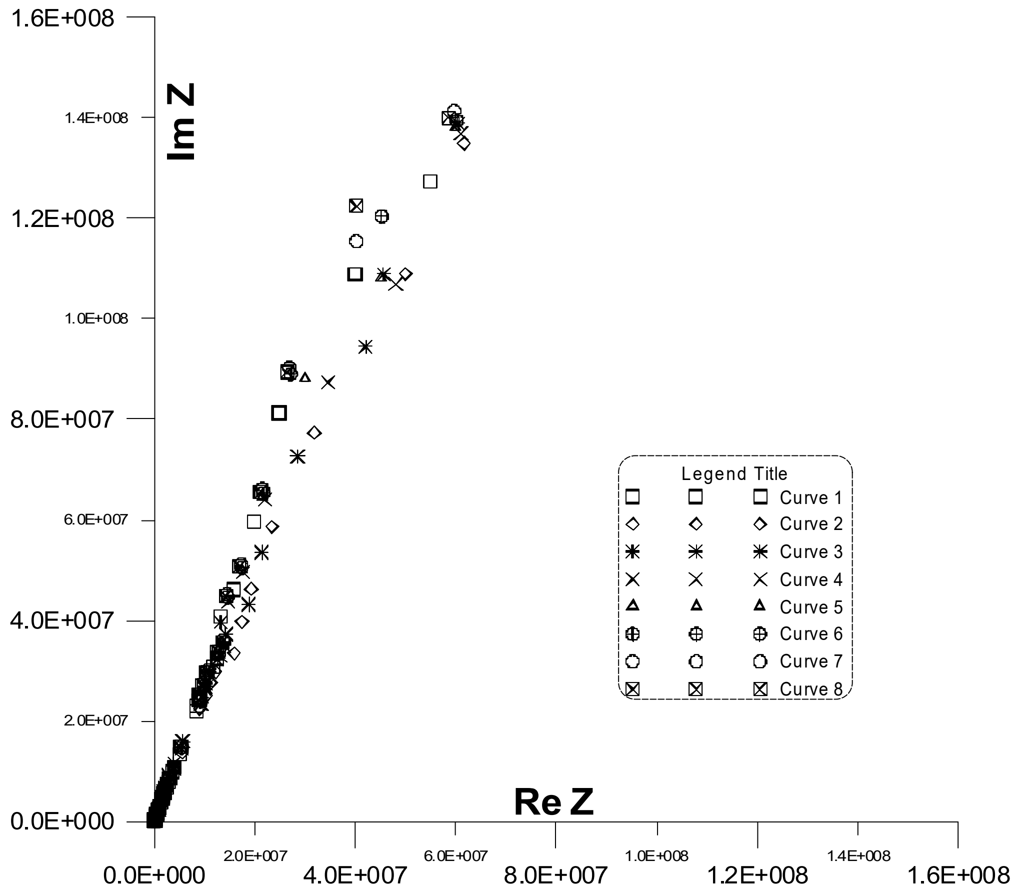


Figure 5. shows the complex impedance of the eight Granite samples.

4. Conclusions

Electrical measurements of some Granite samples were measured at frequencies from 100 Hz up to 10^7 Hz at room temperature (~25^0 C). Samples have a poor primary permeability and grain size in the order of ~0·1 to 0·5 mm with randomly oriented grains. Conductivity depends on the geological and climatic environment of the material origin and upon the direction of the bedding. Minor differences in the composition may lead to a great change in the electrical conductivity. There is no DC conductivity at samples, as detected from conductivity and dielectric constant measurements. With the increase of frequency, the conductivity increases. The dielectric constant values decrease up to nearly 10 kHz and then it settles down after that frequency. The differences in the electrical properties between the samples are small and can be neglected. This is a clear indication that the samples are homogeneous and may have nearly the same composition with different ineffective texture variations. The sample's behavior obeys Jonscher's law. The increase of frequency changes the transport and accumulation of charge carriers. The dielectric loss decreases due to the loss of energy due to the friction of free charges and the increase of hopping between different particles. The present work may be used as a fingerprint for the characterization of Granite rocks.

5. Conflict of Interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

6. Data Availability Statement

The authors confirm that the data supporting the findings of this study are available within the article.

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